

# Past Atmospheric Deposition of Metals in Northern Indiana Measured in a Peat Core from Cowles Bog

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■ A peat core from a calcareous fen was used to assess past metal accumulation from atmospheric sources in northern Indiana. Total concentrations of Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sr, and Zn were measured at intervals along the core, which were dated by  $^{210}\text{Pb}$ , radiocarbon, and pollen analysis. The deposition of airborne metal particulates rose dramatically from presettlement levels as industrialization occurred upwind, with accumulation rates for some metals increasing by 2 orders of magnitude. Recent accumulation rates are about half of the peak rates of the 1970s, presumably because of emission controls and reduced production. This study, the first such record from a calcareous fen, should be less affected by postdepositional mobility than records from acidic peatlands. This method of retrospective, long-term monitoring of airborne particulates can be applied in many areas lacking such records.

## Introduction

Cowles Bog is a raised calcareous fen located in northern Indiana along the south shore of Lake Michigan (Figure 1). It is situated downwind from the city of Chicago and the industrial complexes of Gary and East Chicago and receives a large quantity of airborne particulates. One of the largest American steel mills (Bethlehem Steel, Burns Harbor plant) and a coal-fired power plant [Northern Indiana Public Service Co. (NIPSCO), Bailey plant] operate within 2 km of Cowles Bog.

Previous studies have demonstrated that a record of past metal deposition, especially lead, may be obtained from lake or pond sediments (1-4) or peat profiles from bogs (5-8). Airborne particulates containing metals enter wetlands either directly from atmospheric deposition or indirectly from stream flow after deposition within the watershed. Because some metals are also released through natural weathering processes and carried in streams, this source can complicate stratigraphic records in areas where the degree of erosion or sedimentation has fluctuated. A good site for a study of past metal accumulation would either be one where the watershed has not undergone human-induced change (rare in industrialized America) or one that is not affected by surface runoff.

Cowles Bog is an ideal site for this type of study. It is not fed by any surface streams (9), eliminating this complex source of metal input. Instead, it is a spring mire that is supported by precipitation and groundwater seepage from alkaline artesian flow from below. Peat pore waters near the surface have a pH of 7.16 and an alkalinity of  $9.58 \pm 1.64$  mequiv  $\text{L}^{-1}$  (10). As a result, metal deposition is

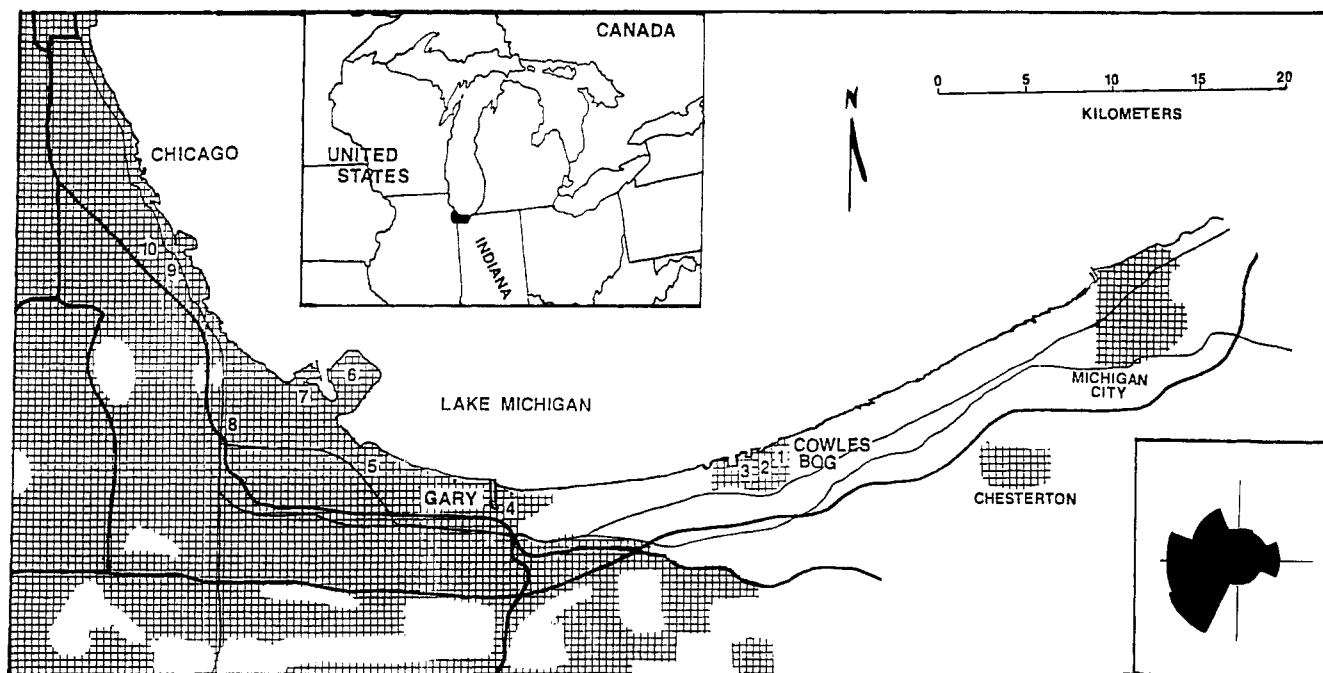
from airborne particulates and precipitation, and metal accumulation at this site should be a good indication of local atmospheric deposition. The sample site is  $>500$  m from the nearest road and thus indicates ambient atmospheric deposition rather than local output from automobile exhaust (11, 12). This is the first such study from a calcareous fen, and the first study of this type in the Great Lakes region.

## Methods

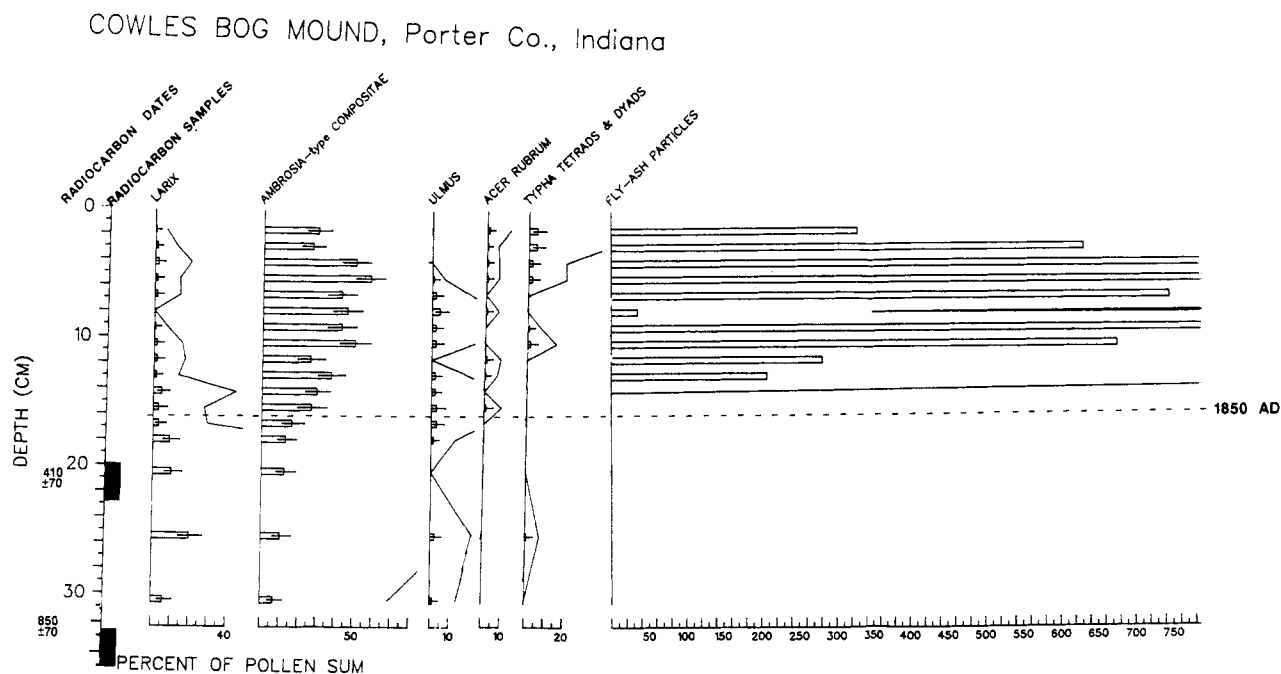
Two sediment cores were taken 10 m apart on a *Sphagnum* mound in the center of Cowles Bog with 10- and 15-cm-diameter PVC tubes cut to 1.5-m lengths. The tubes were pushed into the peat to a depth of  $\sim 0.5$  m. The cores were retrieved by digging trenches adjacent to the tubes, rotating them lengthwise, and lifting them out sideways in order to prevent any loss of the sediment column. This type of device provides abundant sample, retrieves the sediment with minimal distortion, and is inexpensive. The tubes were then cut open to remove the sediment cores. The 15-cm core was sectioned into 1.25-cm wafers and subdivided into samples for radiocarbon dating, pollen analysis, metal analysis, and  $^{210}\text{Pb}$  dating. Sections of the 10-cm core were packed into 33-cm<sup>3</sup> beakers, weighed, oven-dried, and reweighed to calculate mass/volume.

The sedimentation rate was determined through a combination of  $^{210}\text{Pb}$  dating, radiocarbon dating, and pollen stratigraphy. Two radiocarbon dates were obtained on peat from the 15-cm core. This large-diameter core provided adequate material for sampling every 2.5 cm of depth, allowing accurate time-stratigraphic resolution. A deeper date was obtained from a vibracore sample (13), taken within 10 m of the other cores.

The  $^{210}\text{Pb}$  chronology of the last 150 years was determined through the analysis of samples from 18 depth intervals.  $^{210}\text{Pb}$  was measured through its granddaughter product  $^{210}\text{Po}$  with  $^{208}\text{Po}$  added as an internal yield tracer. The polonium isotopes were distilled from 2 g of dry sediment at 550 °C following pretreatment with concentrated HCl and plated directly (without  $\text{HNO}_3$  oxidation) onto silver planchettes from a 0.5 N HCl solution (modified from ref 14). Activity was measured for  $(1-3) \times 10^5$  s with Si-depleted surface barrier detectors and an Ortec Adcam alpha spectroscopy system. Unsupported  $^{210}\text{Pb}$  was calculated by subtracting supported  $^{210}\text{Pb}$  (estimated from asymptotic activity at depth) from total activity at each level. Dates and sedimentation rates were determined



**Figure 1.** Location map showing Cowles Bog. Grid pattern indicates industrial/metropolitan areas; lines represent major highways. Numbers locate major industrial plants: (1) Northern Indiana Public Service Co. (NIPSCO), Bailey Generating Station; (2) Bethlehem Steel, Burns Harbor plant; (3) Midwest Steel; (4) USX, Gary Works; (5) NIPSCO, Mitchel Generating Station; (6) Inland Steel; (7) LTV Steel; (8) AMOCO refinery; (9) State Line Generating Station; (10) USX, Southworks. Regional wind patterns are demonstrated by the wind rose from Ft. Wayne, IN, in the lower right corner.



**Figure 2.** Selected pollen taxa extracted from the Cowles Bog core. The dashed line between 15 and 16 cm corresponds to the approximate time of European clearing of farmland between 1845 and 1865 A.D. Radiocarbon ages are shown at left. Reference 18.

according to the crs (constant rate of supply) model (15), with confidence intervals calculated by first-order error analysis of counting uncertainty (16).

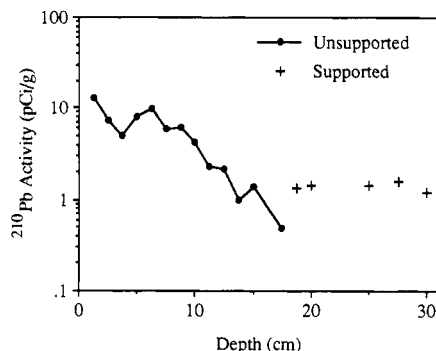
Pollen samples were removed at 1.25-cm intervals by inserting sections of plastic pipet tubing into the core so that each sample contained approximately 1 cm<sup>3</sup> of sediment. These samples were then treated by standard pollen extraction procedures (17, 18).

Metal concentrations were determined at the Analytical Services Laboratory at Michigan Technological University. Two (2- and 3-gm) samples from each of 20 depth intervals were hand-ground with an acid-washed mortar and pestle

and digested by a HClO<sub>4</sub> method. This digestion procedure dissolves all but the most resistant silicate minerals and is quite effective in extracting trace metals from peat (19). Organics were removed by heating each sample in 20 mL of HNO<sub>3</sub> until the foaming stopped. HClO<sub>4</sub> (10 mL) was then added, and the samples were heated until fumes were observed. To dissolve the salts, 10 mL of HCl was added, heated to a boil, filtered, and diluted to 100 mL with deionized water. Blank samples were made by the same method, excluding the peat sample. The solutions were analyzed for Al, Cd, Cr, Co, Cu, Fe, Pb, Mn, Ni, Sr, and Zn by flame atomic absorption spectroscopy using

**Table I. Historical Developments in the Vicinity of Cowles Bog**

Vegetational Changes	
1840–1855	logging, land clearance, invasion of <i>Ambrosia</i> into farmlands, invasion of <i>Acer rubrum</i> into swamp
early 1900s	agricultural ditching of the Cowles Bog Wetland Complex (23)
1960–1975	<i>Ulmus</i> populations severely reduced by Dutch Elm disease (22)
1961–1970	<i>Typha</i> coverage increases from 3.5 ha to 9.7 ha in Cowles Bog (23)
1970–1975	<i>Typha</i> coverage increases from 9.7 ha to 32.3 ha in Cowles Bog (23)
Industrial Developments	
1850–1852	completion of the Buffalo and Mississippi Railroad (1 km south), Michigan Central Railroad (3 km southeast) and Michigan Southern Railroad (2 km southwest) (21)
1902	production begins at Inland Steel plant in East Chicago, 35 km west of Cowles Bog (27)
1905	production begins at United States Steel in Gary, 18 km west of Cowles Bog (27)
1907–1908	fire recorded by a fire scar on <i>Pinus strobus</i> 12 m from core location
1962	power production begins at Northern Indiana Public Service Co., Bailey Power plant, 2 km west of Cowles Bog
1969	coke production begins at Bethlehem Steel, Bailey plant, 2 km west of Cowles Bog



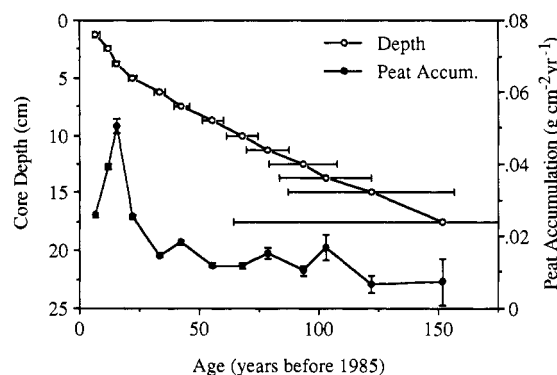
**Figure 3.** Lead-210 activity vs depth for Cowles Bog. Unsupported  $^{210}\text{Pb}$  is calculated by subtracting mean supported activity from total  $^{210}\text{Pb}$  at each level.

element-specific hollow cathode lamps.

## Results

**Dating the Core.** The pollen analysis (Figure 2) showed several changes, which can be correlated with historical developments near the study site (Table I). The increase of ragweed (*Ambrosia*) pollen near a depth of 15 cm indicates the clearing of land by European settlers, which occurred in this region between 1840 and 1855 (18, 20, 21). Also, the decrease of elm (*Ulmus*) pollen between 5.0 and 3.75 cm results from the spread of Dutch Elm disease, which occurred regionally between 1960 and 1975 (22). The postsettlement disturbances to the wetland from logging, fire, and drainage are apparent by the decrease of tamarack (*Larix*) and the subsequent invasion of red maple (*Acer rubrum*) and cattail (*Typha*) (23). Postsettlement industrialization is evident from the accumulation of microscopic particulate matter identified as fly ash (2).

Lead-210 activity decreases downcore in a nonexponential manner, reaching supported values ( $1.38 \pm 0.05 \text{ pCi g}^{-1}$ ) below 18 cm (Figure 3). The unsupported activity profile shows a marked kink around 4 cm, indicating a major change in peat accumulation that can only be fully explored by the crs dating model. The cumulative unsupported  $^{210}\text{Pb}$  in this profile is  $13.03 \text{ pCi cm}^{-1}$  and the equivalent  $^{210}\text{Pb}$  flux ( $0.42 \text{ pCi cm}^{-2} \text{ yr}^{-1}$ ) is very close to the estimated mean deposition rate for atmospheric fallout (24). The age/depth relationship and peat accumulation profile derived from crs calculations are shown in Figure 4 along with error bars representing 1 standard deviation (SD) propagated from counting uncertainty. The error terms become notably large for dates greater than 100–120 years ( $\sim 5$   $^{210}\text{Pb}$  half-lives), reflecting the uncertainty of estimating the small amount of  $^{210}\text{Pb}$  remaining in older sediments. Net peat accumulation, which is relatively constant before ca. 1960 ( $0.007\text{--}0.018 \text{ g cm}^{-2} \text{ yr}^{-1}$ ) rises dramatically within the last two decades. However, this



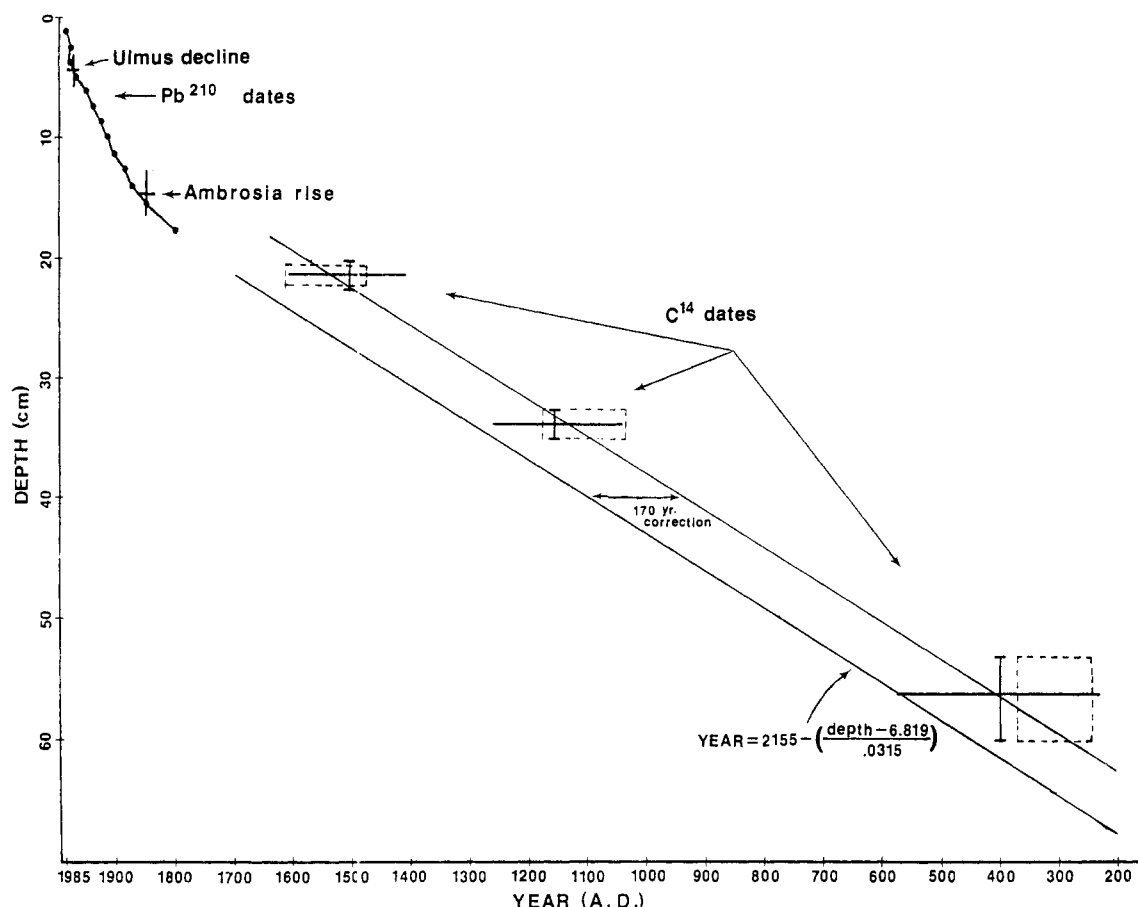
**Figure 4.** Age/depth relationship and peat accumulation for the uppermost 18 cm of the Cowles Bog core, as calculated by the crs model. Error bars represent 1 SD propagated from counting uncertainty.

increase probably represents the incomplete humification of the uppermost peat, rather than any increase in the rate of peat growth.

Radiocarbon dates of older strata yielded ages of  $410 \pm 70 \text{ yr B.P.}$  at 20–22.5-cm depth (GX-11494),  $850 \pm 70 \text{ yr B.P.}$  32.5–35-cm depth (GX-11492), and  $1640 \pm 75 \text{ yr B.P.}$  at 53–60-cm depth (GX-11493). Conversion of these dates to reflect calendar years rather than radiocarbon years, and noncounting uncertainty in dating (25), resulted in minor corrections to the two older dates, and a larger correction to the younger date (Figure 5). The converted 95% confidence limits on the youngest date are large because of variations in atmospheric  $^{14}\text{C}$  during this time period (26). Whether expressed in radiocarbon years or calendar years, regressions through the three radiocarbon dates indicate that they are internally consistent ( $R^2 = 0.99$ ), suggesting that sediment accumulation rates remained fairly constant throughout the interval dated by radiocarbon.

The three dating methods agree favorably with one another (Figure 5). The pollen and  $^{210}\text{Pb}$  chronologies support each other during the postsettlement time period. *Ambrosia* pollen, expected to increase between 1840 and 1855 (18), rises to exceed twice its presettlement background value between 1857 and 1866 (14.5–15.5 cm) according to the  $^{210}\text{Pb}$  chronology. *Ulmus* pollen, expected to decrease between 1955 and 1970 (22), is reduced below half its prior average between 1947 and 1968 (4.5–5.5 cm) according to the  $^{210}\text{Pb}$  chronology.

The radiocarbon dates agree fairly well with the other dates but may be slightly too old because of the incorporation of “dead” carbon from groundwater carbonates upwelling from beneath this calcareous fen. Extrapolation of the trend in sediment accumulation between the radiocarbon dates to the youngest  $^{210}\text{Pb}$  date shows a dis-



**Figure 5.** Sediment age vs depth from Cowles Bog. Lead-210 dates are shown with the location of the strata dated by the decrease of fossil elm (*Ulmus*) pollen and the increase in fossil ragweed (*Ambrosia*) pollen. Raw radiocarbon dates with standard deviations (dashed boxes) have been converted to calendar years (25) and are displayed with their 95 % confidence limits (error bars). The least-squares regression through the radiocarbon dates is shown with a lower (younger) regression showing the trend "corrected" for 170-year error due to input of dead carbon from groundwater. Note that the correction allows for a smooth transition between the  $^{210}\text{Pb}$  chronology and the  $^{14}\text{C}$  chronology but does not change the sediment accumulation rate below the top radiocarbon date or above the lowest lead date.

parity of 170 yr (Figure 5). This disparity can be reconciled by assuming that (1) the lowermost  $^{210}\text{Pb}$  date is correct, (2) the 170-yr disparity between the radiocarbon trend and the lowermost  $^{210}\text{Pb}$  date is due to contamination from groundwater carbonate, and (3) groundwater contamination from carbonate remained similar throughout the time period covered by the radiocarbon dates. As a result, a better approximation of the sediment age can be obtained by moving the age trend between radiocarbon dates forward 170 years. This minor adjustment in age allows the calculation of sediment accumulation rates for the three samples between the oldest  $^{210}\text{Pb}$  date and the youngest radiocarbon date (1656–1815 A.D.), but does not affect the values obtained for the sediment accumulation rate below the youngest radiocarbon date or above the oldest  $^{210}\text{Pb}$  date.

A general trend of past low sediment accumulation rates gradually increasing to higher recent accumulation rates is evident in both the  $^{210}\text{Pb}$  chronology and the composite chronology (Figure 5). This apparent increase in peat accumulation probably is an artifact of decaying organic matter. As time goes on, the organic matter representing a span of time should become more decayed and compacted, giving the appearance of a lower rate of accumulation in older strata.

**Metal Content and Accumulation.** Metal concentration profiles are shown in Figure 6. These data were converted to rates of metal accumulation (Figure 7) by multiplying by the sediment accumulation rate for each interval. The accumulation rates for intervals above 18

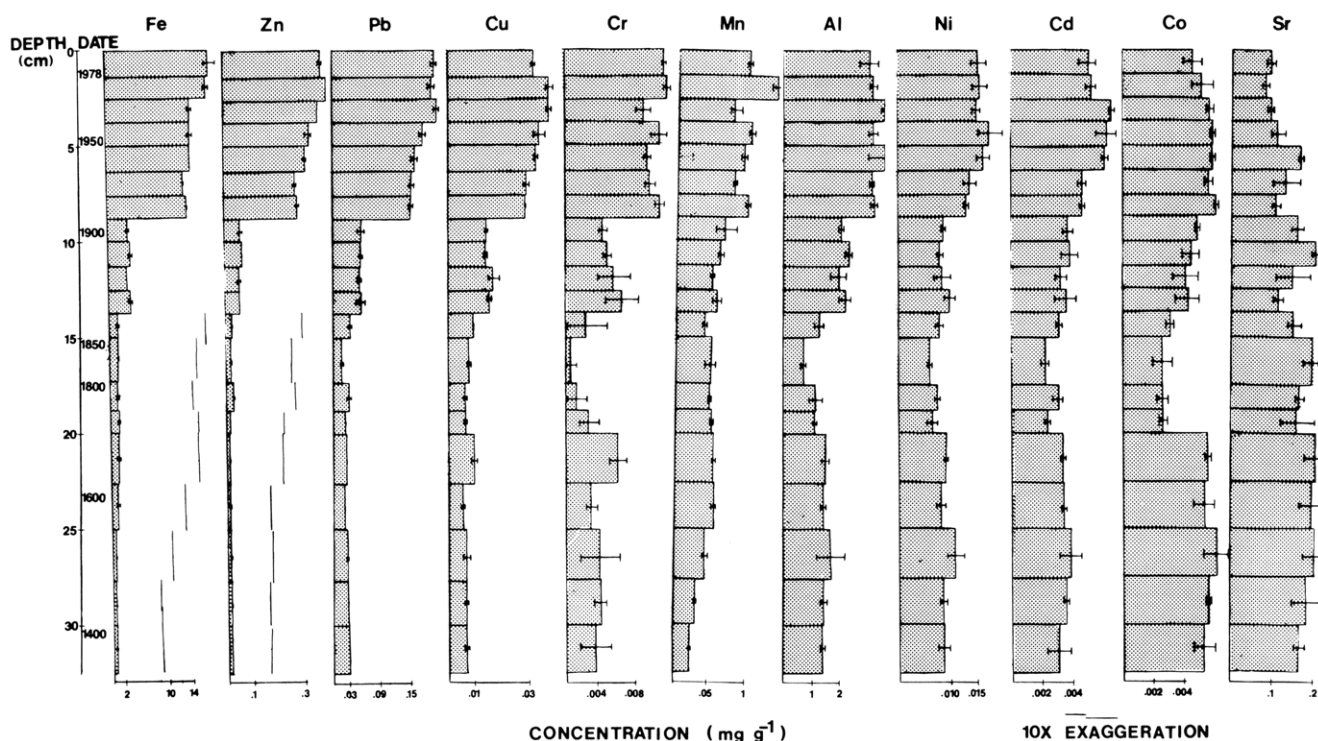
**Table II. Presettlement Metal Accumulation Rates (1339–1656 A.D.) and Peak Accumulation Rates 1970–1973 (72) or 1973–1978 (76) at Cowles Bog ( $\text{mg m}^{-2} \text{yr}^{-1}$ )<sup>a</sup>**

	presettlement accum	peak accum
Al	$58.4 \pm 5.1$	1632.5 (72)
Cd	$0.121 \pm 0.010$	2.79 (72)
Co	$0.241 \pm 0.020$	3.02 (72)
Cu	$0.266 \pm 0.018$	20.83 (72)
Cr	$0.152 \pm 0.025$	4.70 (76)
Fe	$41.65 \pm 8.72$	7430 (72)
Pb	$1.079 \pm 0.065$	101.9 (72)
Mn	$17.15 \pm 5.66$	553.9 (76)
Ni	$0.371 \pm 0.052$	7.73 (72)
Sr	$8.060 \pm 0.981$	57.0 (72)
Zn	$0.802 \pm 0.035$	210.4 (72)

<sup>a</sup>The numbers shown for presettlement accumulation are the mean and standard deviations of the pre-1656 samples.

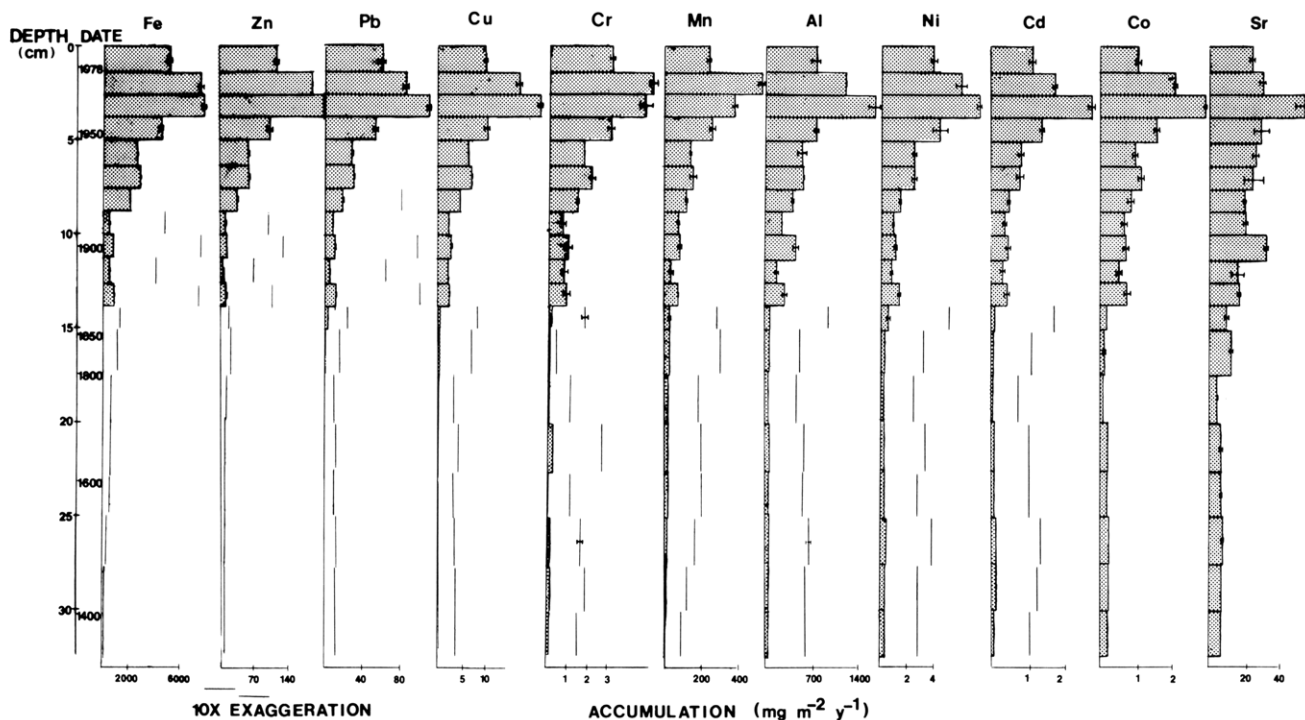
cm were calculated by using the  $^{210}\text{Pb}$  chronology, while the lower interval rates were obtained from the regression of the three radiocarbon dates. The radiocarbon dates provided an average sediment accumulation rate of  $0.00419 \text{ gm cm}^{-2} \text{yr}^{-1}$  for the period between 465 A.D. and 1775 A.D. (60–18.75-cm depth). The calculated postsettlement metal accumulation rates are much higher than the presettlement rates (Table II). Peak postsettlement metal accumulation rates range from 700% (Sr) to 26200% (Zn) of the presettlement rates. Post-1977 accumulation rates (the top-most sample) are about half the peak accumulation values recorded in the 1970s.

# COWLES BOG metal concentration ( $\text{mg g}^{-1}$ )



**Figure 6.** Metal concentrations vs depth in Cowles Bog. Metals are arranged from left to right depending upon the percentage increase from presettlement values to peak values. Error bars show the range between two samples.

# COWLES BOG metal accumulation ( $\text{mg m}^{-2} \text{y}^{-1}$ )



**Figure 7.** Rate of metal accumulation vs time in Cowles Bog. Accumulation values were calculated by multiplying concentration values by sediment accumulation values (Figure 3). The thin line above low values represents a 10X exaggeration. Error bars show the range between two samples.

The metals are ranked from left to right in Figures 6 and 7 according to their percentage increase in accumulation rate over background levels. Fe and Zn increase the greatest amount, reaching peak accumulation rates of 178 and 262 times their presettlement background rates, and showing large increases in concentrations between 1928 and

1942 A.D. Pb, Cu, and Cr reach peak accumulations of 94, 78, and 31 times their presettlement values and also show abrupt increases in concentration between 1928 and 1942. These metals probably reflect the input of fly ash from the regional development and expansion of the steel industry. The parallel increase in fly ash (Figure 2) implies

that much of the anthropogenic metal burden in Cowles Bog is in the form of refractory particulates and, thus, should be resistant to postdepositional translocation within the core. The stratigraphic signals represent historical deposition and are not a diagenetic feature.

The other elements tested had lower increases over presettlement accumulation values, ranging from 32 (Mn) to 7 (Sr). Strontium had the lowest increase in accumulation and even a reverse trend in concentration values. It is probably not a major component of the atmospheric deposition, and much of its input may be from the local upwelling groundwater (9) rather than anthropogenic sources.

The fact that most of the metals analyzed show very similar stratigraphic patterns is somewhat unsettling. Metals derived from different sources might be expected to show less synchronicity than is exhibited here. Part of the problem may be that our sampling interval (1.25 cm) is too coarse to discern slightly different trends in metal emissions. However, it should be noted that the maximum rates of metal accumulation from 2.5 to 3.75 cm correspond to a peak in peat accumulation rates (Figure 4). Since atmospheric metal deposition and peat growth should be independent of one another, it is possible that these maxima reflect an anomaly in dating or peat accumulation. This contention is supported by the fact that Sr, which is mostly nonanthropogenic, also reaches a maximum at this level. This anomalous peak in Sr occurs only at this level, however, and the profile is otherwise complacent. Thus, the accumulation trends for anthropogenic metals may be slightly distorted by anomalously high peat accumulation for this single sample (2.5–3.75 cm), but should otherwise provide a fairly accurate record of metal deposition at Cowles Bog.

## Discussion

**Chronology of Industrial Development.** The industrial development of the Chicago/northwestern Indiana region is reflected in the metal accumulation chronology. The first agricultural/residential growth within the area was aided with completion of the Michigan Central Railroad and the Michigan Southern Railroad (both later absorbed by New York Central) through the area in 1852 (21, 27). The Baltimore and Ohio Railroad line was completed in 1874. The first rise in metal accumulation over presettlement levels (1850–1890) is probably related to coal burning on steam locomotives, residential fires, and eventually the iron and steel industry developing in the region.

In 1902 and 1908 production began at what were to become the giant steel complexes of Inland Steel and United States Steel in East Chicago and Gary (27) (Figure 1). Industrial development and production increased throughout the first half of this century. This industrialization is reflected in the increased metal accumulations between 1900 and 1960, particularly the sharp increase occurring between 1928 and 1942.

The next large increase in metal accumulation at Cowles Bog, between 1962 and 1969, was probably related to the first operation of the NIPSCO Bailey Power Station and the giant Bethlehem Steel Burns Harbor plant 2 km from the core site. The coal-fired power plant began operations in 1962, while the steel mill started operation of its coke batteries in 1969. Peak accumulation values are found in the two samples dated between 1969 and 1978. The last sample, postdating 1978, shows lower values, which are most likely a result of lessened production or emission controls.

**Table III. Metal Concentrations and Accumulation Reported from the Cowles Bog Region**

	Cowles Bog <sup>a</sup>	East Chicago <sup>b</sup>	S. Lake Michigan <sup>c</sup>
<b>Pb</b>			
metal concn, mg g <sup>-1</sup>			
top 2.5 cm	0.196	0.463	0.10–0.16
top 14 cm	0.128	0.194	
accum rate, mg m <sup>-2</sup> yr <sup>-1</sup>	81.7 (1973–1978)	81.5 (1975–1976)	13.0 (1972)
<b>Cd</b>			
metal concn, mg g <sup>-1</sup>			
top 2.5 cm	0.00437	0.0104	
top 14 cm	0.00406	0.0049	
accum rate, mg m <sup>-2</sup> yr <sup>-1</sup>	1.87 (1973–1978)	0.82 (1975–1976)	
<b>Zn</b>			
metal concn, mg g <sup>-1</sup>			
top 2.5 cm	0.438	2.46	
top 14 cm	0.330	1.00	
accum rate, mg m <sup>-2</sup> yr <sup>-1</sup>	189 (1973–1978)	98.4 (1975–1976)	
<b>Cu</b>			
metal concn, mg g <sup>-1</sup>			
top 2.5 cm	0.0364	0.119	
top 14 cm	0.0294	0.045	
accum rate, mg m <sup>-2</sup> yr <sup>-1</sup>	16.55 (1973–1978)	16.5 (1975–1976)	

<sup>a</sup>This study. <sup>b</sup>Reference 28. <sup>c</sup>Reference 1.

**Comparison to Other Regional Records.** The metal concentrations and accumulation rates recorded from the upper layers of Cowles Bog are similar to other studies within the region (Table III). The peat concentrations of Pb, Cd, Zn, and Cu are slightly less than those recorded in soils from nearby East Chicago (28), but the mid-1970s' accumulation rates are remarkably similar. The modern (1972) deposition of lead in southern Lake Michigan has been estimated at 13 mg m<sup>-2</sup> yr<sup>-1</sup> (1). The rate calculated from this study (50–107 mg m<sup>-2</sup> yr<sup>-1</sup>) is higher, as might be expected since Cowles Bog is closer to the source of emissions (29).

An important contribution of this study is the estimation of "natural" or background fluxes of metals into regional watersheds. These values, representing average metal accumulations from 1350 to 1650 A.D. (Table II) can be used as a regional background for evaluating changes in accumulation occurring since settlement.

## Conclusions

The metal concentrations and calculated accumulation rates from Cowles Bog show a dramatic increase during the last 130 years, especially for the elements Fe, Zn, Pb, Cu, and Cr. These calculated accumulation rates are consistent with the regional history of industrial development. The values obtained in this study can be used to estimate preindustrial metal accumulation rates and demonstrate the timing and magnitude of increases due to industrialization. These accumulation values can be extrapolated to estimate future soil metal concentrations.

A calcareous fen, such as Cowles Bog, provides an ideal archive for the history of metal deposition because it is not subject to inputs of clastics from streams or to the problems of sediment redistribution that are characteristic of lacustrine environments.

## Acknowledgments

The metal analyses were conducted by Mary Hillebrand and Loren Severs at the Analytical Services Laboratory of Michigan Technical University. Several figures were drafted by Kenneth Klick. Wanda Manek assisted with the coring of the bog. Douglas Wilcox, Lou Brenan, and Todd Thompson provided consultation on metal pollution and the stratigraphy of Cowles Bog.

**Registry No.** Al, 7429-90-5; Cd, 7440-43-9; Co, 7440-48-4; Cu, 7440-50-8; Cr, 7440-47-3; Fe, 7439-89-6; Pb, 7439-92-1; Mn, 7439-96-5; Ni, 7440-02-0; Sr, 7440-24-6; Zn, 7440-66-6.

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Received for review July 5, 1989. Revised manuscript received November 15, 1989. Accepted December 15, 1989. This research was funded by the National Park Service.

# Atmospheric Methane: Recent Global Trends

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■ We report globally averaged concentrations of atmospheric methane for every month of the past 8 years based on measurements taken at six locations ranging in latitude from within the Arctic Circle to the South Pole. This record shows that methane concentration increased at an average rate of  $16.6 \pm 0.4$  ppbv/yr or  $\sim 1.02 \pm 0.02\%$ /yr over 8 years. This trend has not been constant according to our record but has varied between  $12 \pm 2$  and  $23 \pm 2$  ppbv/yr over 2-year periods after seasonal variations are removed. The causes of these interannual variations are not known. We also show that the total mass of methane in the earth's atmosphere undergoes seasonal variations, with highest levels during late fall and early winters of the northern hemisphere and lowest levels in the summers. After the main features of the record are taken into account, residual random fluctuations remain, which have a variability of only 3 ppbv or 0.2% of the mean concentrations. The variation of the trends at different times during the past decade accounts for the differences of trends reported in various studies. Uncertainties in our estimates are reported as 90% confidence limits.

## 1. Introduction

There is no longer any doubt that the global concentration of atmospheric methane is increasing (1-6). In the

future, continued increases may cause global warming and affect the atmospheric cycles of carbon monoxide, ozone, and water vapor (see ref 7).

To study the cycle of methane and the causes of increasing trends, intensive and systematic measurements have been taken all over the world during the last decade. Although every experiment has shown that methane is increasing, there have been apparent discrepancies on the rate of increase. Our earliest results suggested trends of nearly 2%/yr (8); a few years later the increase was estimated at 1.3%/yr (4); the longer term measurements of Blake and Rowland (6) showed trends of 1%/yr, and Steele et al. (9) reported even slower increases of slightly less than 0.8%/yr. We have two objectives in writing this paper. The first is to report the average concentrations and trends from our global measurements taken every week at six locations ranging in latitudes from the Arctic to the Antarctic. To our knowledge there are no other records with such frequent global measurements that extend over nearly a decade. From this record we estimate the globally averaged concentrations of methane from every month of the 8 years between September 1980 and September 1988. The second objective is to show that the trends have not been constant over this period but have varied considerably. These variations explain the differences in the trends reported by Khalil and Rasmussen,